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BUOYANT-THERMOCAPILLARY INSTABILITIES OF DIFFERENTIALLY HEATED LIQUID LAYERS

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Abstract

The stability of buoyant-thermocapillary-driven flows in a fluid layer subjected to a horizontal temperature gradient is analysed. Our purpose is the modelization of recent experimental results obtained for a fluid of Prandtl number $Pr=7$, by Daviaud and Vince [Phys. Rev. E, 4432 (1993)] who observed a transition between traveling waves and stationary rolls when the height of fluid is increased. Our model takes into account several effects which were examined separately in previous studies. The relative importance of buoyancy and thermocapillarity is controlled by the ratio W of Marangoni number to Rayleigh number. The fluid layer is bounded below by a rigid plane whose temperature varies linearly along the direction of the thermal gradient. A Biot number is introduced to describe heat transfer at the top free surface. Our stability analysis establishes the existence of a transition between stationary and oscillatory modes when W exceeds a value W_0 which is function of the Biot number. Moreover, two types of oscillatory modes have been identified which differ by the range of variation of their critical parameters (wave number, frequency, angle of propagation) versus W .

I. INTRODUCTION

The dynamical behavior of flows driven by a horizontal temperature gradient in fluid layers bounded above by a free surface has attracted the attention of researchers since many years. The interest in this field has been initiated by the need for understanding the hydrodynamical aspects of crystal growth in a low-gravity environment [1]. In solidification processes like the floating zone technique the existence of thermocapillary convection cells in the melt has been demonstrated experimentally [2]. It was shown later that the primary flow becomes unstable towards oscillatory instability [3]. The instability mechanisms were identified by Smith and Davis [4] in their linear stability analysis of thermocapillary flows. They reported on two classes of instabilities which differ by the energy transfer mechanism. The first mode is a thermal instability which takes the form of either longitudinal rolls or hydrothermal waves. The second mode is closely related to free-surface deformations and involves mechanical transfer of momentum. The stability analysis performed in [4] was restricted to liquid layers of infinite horizontal extent bounded below by an adiabatic rigid surface. An analytical treatment of the stability problem lies on the assumption of a plane-parallel basic flow, an approximation valid far from the vertical walls confining the fluid in the horizontal direction. Steady thermocapillary flows in bounded geometries are two-dimensional with the surface flow turning around near the end walls and recirculating below. Asymptotic matching theories have been used with the lubrication approximation to describe two-dimensional flows confined within a slot [5, 6].

More recently, the coupling between thermocapillarity and buoyancy has been considered through different approaches. The stability of buoyant-thermocapillary-driven flows in unbounded geometries has been addressed in [7] for low-Prandtl-number fluids and in [8, 9] for larger values of the Prandtl number up to $Pr=10$. The analyses performed in [8] and [9] differ by their choice of thermal boundary conditions on the horizontal surfaces. In the case of conducting surfaces, Gershuni et al. [8] yield the result that three-dimensional disturbances in the form of stationary longitudinal rolls are the preferred mode of instability for $Pr>1$. In the case of adiabatic surfaces, Parmentier et al. [9] found that unsteady disturbances in the form of oblique waves are the most unstable.

Two-dimensional buoyant-thermocapillary-driven flows confined within a shallow cavity have been investigated numerically in [10] for low-Prandtl-number fluids and various values of the aspect ratio. These numerical sim-

ulations have shown the existence of complex flow patterns in the form of multicellular steady states and the transition to oscillatory convection has been observed. The onset of oscillatory convection has been carefully analysed in [11] showing that a transition to unsteady flows occurs provided the Grashof number exceeds a lower bound value. Experiments and numerical simulations have been carried out by Villers and Platten [12] for $Pr=4$ corresponding to acetone as the working fluid. Their results confirm the existence of different steady states corresponding to either a monocellular flow or a multicellular flow.

A two-dimensional formulation of the problem aims at modeling experimental results obtained in cavities whose horizontal extent in the direction of the thermal gradient is larger than the extent in the spanwise direction as it is the case in [12, 13]. However, it fails to reproduce the onset of three-dimensional disturbances in extended geometries. To avoid sidewall effects, an annular gap configuration has been used in [14]. Thus, two-dimensional simulations in enclosed cavities and stability analysis of unbounded fluid layers provide two complementary approaches.

Our own interest in the field was motivated by recent experimental results obtained by Daviaud and Vince [15] in a new geometrical configuration. The system they considered was specially designed to study the dynamics of one-dimensional traveling waves. It consists of a long and narrow rectangular container filled with silicon oil and submitted to a horizontal temperature gradient perpendicular to its large extension. The two control parameters of the system are h , the height of fluid and DT , the horizontal temperature difference between the side walls. Depending on the height of fluid, two regimes of instability have been observed when DT is increased above a threshold value DT_c . For small heights, oblique waves propagating in a direction nearly perpendicular to the thermal gradient are the preferred instability patterns, while for large heights, stationary longitudinal rolls are the critical modes.

The existence of stationary or oscillatory modes was already predicted by several authors although it was under quite different conditions. In their analysis of pure thermocapillary flows bounded by two adiabatic horizontal surfaces Smith and Davis [4] have found that stationary longitudinal rolls are the preferred modes for $Pr>1.6$, when a linear base flow profile is considered. In a layer bounded by conducting horizontal surfaces where the primary flow is driven by both thermocapillarity and buoyancy, Gershuni et al. [8] have shown the existence of stationary rolls for $Pr>1$. However, neither the as-

sumption of a linear flow nor the choice of a conducting upper free surface are appropriate to fit the experimental conditions of Ref. 15. When a return basic flow is considered, Smith and Davis [4] always found that instability occurs in the form of oblique hydrothermal waves with an angle of propagation $Y \approx 27^\circ$ for $Pr=6.8$. With the same choice of thermal boundary conditions, Parmentier et al. [9] have shown that the value of Y varies when buoyancy effects are taken into account. In the particular case $Pr=7$, these authors have found two types of critical oscillatory modes which differ by the value of their angle of propagation. For low values of the Rayleigh number they recovered the oblique waves of Smith and Davis [4] with $Y \approx 30^\circ$, while increasing the Rayleigh number the angle of propagation undergoes a radical change and takes larger values. Instability in the form of longitudinal rolls ($Y=90^\circ$) are predicted to occur when the critical Rayleigh number is twice the critical Marangoni number. Still increasing Ra , the angle of propagation decreases until it reaches $Y=0^\circ$ for $Ra > 4.103$. Thus, the existence of traveling longitudinal rolls ($Y=90^\circ$) was predicted by Parmentier et al. [9], but they never found stationary modes as those observed by Daviaud and Vince [15]. A comparison between earlier theoretical results [4, 8] shows that a key requirement for the existence of stationary modes is the presence of a basic temperature profile which is unstable in the sense of Rayleigh-Bénard or Bénard-Marangoni instability.

The aim of our study is to build a model able to reproduce the transition between oscillatory and stationary modes observed by Daviaud and Vince [15] when the height of fluid is increased. For this purpose a particular attention has been paid to the choice of appropriate thermal boundary conditions at the top and bottom horizontal boundaries.

II. PROBLEM FORMULATION

The experimental set up to which we shall refer has already been described in [15]. It consists in a rectangular container filled with fluid up to a height h . Using cartesian coordinates (x, y, z) with the z -axis along the vertical direction, the dimensions of the container are respectively L_x and L_y following the x and y directions. The aspect ratio $L_y/h^3 \approx 20$ is large so that in our theoretical formulation we shall consider a system with infinite extent along the y -axis. A first set of experiments was achieved with $L_x=1\text{cm}$, thus allowing for aspect ratio variations in the range $1 \leq L_x/h \leq 10$ when the height h is varied. More recently, experiments have been performed with larger values of L_x in order to increase the minimum value of L_x/h up to 3 [16]. However, this value is still too small to allow for the assumption of

an infinite extent along the x-axis in the theoretical description. In our simplified model this difficulty is overcome by neglecting the vertical side walls effects and considering the fluid behavior near mid-width.

The fluid filling the container is incompressible with constant kinematic viscosity ν and thermal diffusivity k . The Prandtl number $Pr=\nu/k$ is kept fixed at the value $Pr=7$, corresponding to the silicon oil used in [15]. The fluid density is temperature dependent according to the law

$$\rho = \rho_0 [1 - \alpha_T (T - T_0)] \quad (1)$$

where α_T is the thermal expansion coefficient and T_0 a reference temperature.

The two opposite lateral boundaries distant of L_x are maintained at different temperatures, T_0 and $T_1 > T_0$ creating a horizontal temperature gradient that drives the fluid upward near the hot wall and downward near the cold wall and generates a whole circulation inside the rectangular cross section (Ox, Oz) of the container.

The fluid layer is bounded at the bottom by a rigid surface while the top boundary is a free surface open to ambient air. Surface tension s which acts at the liquid-air interface admits temperature variations approximated by the linear equation of state

$$s = s_0 - g (T - T_0) \quad (2)$$

where $g = -(ds/dT)$ is a positive constant. Otherwise, we shall assume a flat, non deformable interface which remains at rest. Owing to the horizontal temperature gradient, tangential capillary forces proportionnal to s/x drive the fluid near the interface from the hot regions towards the cold regions, thus reinforcing the thermogravity circulation.

Though the static equilibrium state is broken as soon as the horizontal temperature gradient is applied, it is assumed that in the case of a small driving temperature difference, the circulation is weak and energy transfer is dominated by conduction so that

$$\nabla^2 T = 0 \quad (3)$$

The above equation has to be solved with appropriate boundary conditions on the bottom and top surfaces. In experiments reported in [15] the bottom plate is made of glass whose thermal conductivity is ten times higher than the fluid conductivity but is much less than the conductivity of the vertical sidewalls made of copper. Thus, we shall assume that the bottom wall temperature remains fixed and varying linearly as

$$T_w = T_0 + bT_x.$$

where $bT=(T_1-T_0)/L_x$ is the constant positive horizontal temperature gradient. The thermal boundary condition for the fluid is thus

$$T = T_w \text{ at } z = 0, (4)$$

On the top free surface, it is generally assumed that the heat transfer between liquid and air is conveniently approximated by the following relation [17]

$$f(T;z) + Bi (T - T_a) = 0, (5)$$

where Bi is a Biot number and T_a is the temperature of air at some distance h_a from the interface. In Eq. (5) z is considered as a non dimensional variable, being scaled with h . We shall further assume that $T_a=T_0+bTx$ as it was done in Ref. 4. The determination of the value of Bi , which a priori takes different values whether heat is exchanged by conduction or by convection, is rather empirical. In a purely conductive state Bi can be expressed as

$$Bi = f(ca;cf) f(h;h_a) , (6)$$

where ca and cf are respectively the thermal conductivity of air and fluid. Whereas, in a convective state it has been shown in [17] that the Biot number also depends on the horizontal wavenumber of the convective motion. A Biot number of order one is considered as a good estimate in the experimental conditions of Ref. 15.

A solution for the fluid temperature satisfying Eq. (3) with the boundary conditions (4) and (5) is

$$T = T_0 + bTx (7)$$

The flow induced by the temperature distribution (7) is derived from the governing equations for the fluid layer which express conservation of mass, momentum and energy. Within Boussinesq's approximation these equations are

$$\tilde{f}(u;t) + u \cdot \tilde{N} u = - \tilde{f}(1;r_0) \tilde{N} p + n \tilde{N}^2 u - \tilde{f}(r;r_0) g e_z (8a)$$

$$\tilde{f}(T;t) + u \cdot \tilde{N} T = k \tilde{N}^2 T (8b)$$

$$\tilde{N} \cdot u = 0 (8c)$$

where $u = (u, v, w)$ is the velocity vector, p the pressure, g the gravity acceleration and e_z the upward unit vector in z -direction. The mechanical boundary conditions on the rigid bottom plate are

$$u = v = w = 0 \text{ at } z = 0. (9)$$

On the top free surface the boundary conditions express the impermeability condition and the balance between viscous tangential stress and thermocapillary forces

$$w = 0, (10a)$$

$$h f(u;z) = f(s;x) , (10b)$$

$$h f(v;z) = f(s;y) , (10c)$$

where h is the dynamical viscosity.

Henceforth all the variables will appear in dimensionless form. The scaling are respectively: h for distances, k/h for velocity, h^2/n for time, bTh for temperature and r_0nk/h^2 for pressure.

III. THE BASIC STATES

Neglecting vertical side walls effects, the primary state is assumed to be a plane parallel steady flow with only an x -component depending of the z -coordinate: $u=(U_0(z), 0, 0)$. The interaction of the base flow with the horizontal temperature gradient induces a z -dependent temperature distribution $t_0(z)$. Elimination of pressure in the set of Eqs. (8) leads to the differential equations

$$D^3U_0 = Ra(11a)$$

$$D^2t_0 = U_0(11b)$$

where the notation Dd/dz has been introduced and the Rayleigh number is defined as

$$Ra = f(aTbTgh^4/nk).(12)$$

The associated boundary conditions are

$$U_0 = 0 \text{ and } t_0 = 0 \text{ at } z = 0, (13a, b)$$

$$DU_0 = -Ma \text{ and } Dt_0 + Bi t_0 = 0 \text{ at } z = 1, (13c, d)$$

where the Marangoni number is defined as

$$Ma = f(gbTh^2/hk).$$

The closure of the system is ensured by addition of the zero-mass flux condition through any vertical section

$$i(0;1; U_0 dz) = 0. (14)$$

In the experiments which have motivated this study, the fluid dynamical behavior is controlled by two parameters: the horizontal temperature difference $(T_1 - T_0)$ which measures the strenght of the flow and the height of liquid in the container which gives the relative importance of the buoyancy and thermocapillary effects. The corresponding dimensionless parameters in our theoretical model are the Rayleigh number and the ratio of Marangoni number to Rayleigh number

$$W = f(Ma;Ra) ,$$

which varies like h^{-2} .

Changing the velocity scale by the transformation $U_0(z)=Rau_0(z)$, the solutions for the basic state are

$$u_0(z) = bbc[(f(1;48) (8z^3 - 15 z^2 + 6z) - f(W;4) (3z^2 - 2z)).(15a)$$

$$t_0(z) = f(Ra; 48) z \text{ bbc}[(f(1; 20) (8z^4 - 25z^3 + 20z^2 - 3m) - W (3z^3 - 4z^2 + m))(15b)$$

where $m=f(Bi; 1+Bi)$. Expression (15a) for the velocity profile is the sum of two terms, the first term corresponds to a pure buoyancy-driven flow and the second term stands for the contribution of a pure thermocapillary-driven flow; the resulting profile is shown in Fig. 1(a) for $W=1$. Expression (15b) generalizes those found in [4, 8, 9] when the top and bottom thermal boundary conditions are symmetrical. The particular case of insulating horizontal surfaces examined in [4, 9] corresponds to the value $m=0$, while the case of conducting horizontal surfaces examined in [8] corresponds to the value $m=1$. In the present study where the thermal boundary conditions are different on top and bottom surfaces, one can nevertheless recover the basic states corresponding to symmetrical conditions as limiting cases when Bi_0 and Bi . When Bi , Eqs. (13b) and (13d) reduce to $t_0(0)=t_0(1)=0$, as in the conducting case. When $Bi=0$, Eqs. (13b) and (13d) reduce to $t_0(0)=Dt_0(1)=0$, but due to a property of our model which can be shown by integrating Eq. (11b) and using Eq. (14), we also have $Dt_0(0)=Dt_0(1)$, thus recovering the insulating boundary conditions for the basic temperature profile. However, this property will no longer be true for the temperature disturbances.

To our knowledge, the stability of a buoyant-thermocapillary flow has been investigated for the first time by Laure and Roux [7], their analysis being restricted to low-Prandtl-number fluids ($Pr < 1$). More recently, the range of variation of the Prandtl number was extended up to the value $Pr=10$, by Gershuni et al. [8] in the case of conducting horizontal surfaces and by Parmentier et al. [9] in the case of adiabatic horizontal surfaces. The comparison between the theoretical results obtained in [7, 8, 9] and available experimental data is difficult for several reasons. The main differences between theoretical and experimental approaches are a different geometrical configuration and different thermal boundary conditions. The parameter m related to the Biot number, introduced in the present study provides an additional degree of freedom to the system. Variations of this parameter allow for a gradual change in the thermal boundary conditions which is expected to result in the onset of different modes of instability. In the variation range $0 \leq m \leq 1$, the deformations of the temperature profile are shown in Fig. 1(b) for the two extremal values of m and for an intermediate value. It must be pointed out that $t_0(z)$ which is an increasing function of z when $m=0$, becomes an inverse-S-shaped curve when m increases. In the latter case, there are two sublayers adjacent to the lower and upper surfaces which are potentially un-

stable towards convective and surface tension driven instabilities and which are separated by a stably stratified layer. Among the numerical and analytical studies mentionned above there is only one contribution by Villers and Platten [12] which corresponds to non symmetrical thermal boundary conditions. With a conducting bottom wall and an adiabatic upper free surface, these authors reported on the existence of locally unstable vertical stratifications of the temperature ($dT/dz < 0$).

It is known since the work by Smith [17], that instability mechanisms are different whether the basic-state vertical temperature profile is stabilizing or destabilizing in the sense of Marangoni instability. In [4] the basic-state was such that a destabilizing temperature profile is associated with a “linear flow” and a stabilizing temperature profile is associated with a “return-flow”. In the present study the velocity profile is always a “return-flow” and the temperature profile is either stabilizing or destabilizing depending upon the choice of thermal boundary conditions. The stability analysis of this basic-state is presented in the next section.

IV. LINEAR STABILITY ANALYSIS

The basic state is perturbed in such a way that the velocity, temperature and pressure fields in the resulting state are written as the sum of the base flow variables and a perturbation small enough for the linearization to be a valid procedure

$$u = (U_0 + u_1, v_1, w_1), T - T_0 = x + t_0 + q_1, p = p_0 + p_1, \quad (16)$$

where all the variables are dimensionless. The perturbations are sought under the form

$$\{u_1, v_1, w_1, q_1, p_1\} = \{u(z), v(z), w(z), q(z), p(z)\} \exp i(ax + by + Wt), \quad (17)$$

where a and b are respectively the wave numbers in the streamwise and spanwise directions and $W = w + iv$ is the complex growth rate of the instability. At marginal stability ($v=0$) the z -dependent eigenfunctions are solutions of the following differential system

$$D^2u = Lu + GrwDu_0 + iap \quad (18a)$$

$$D^2v = Lv + ibp \quad (18b)$$

$$D^2w = Lw - q + Dp \quad (18c)$$

$$D^2q = [k^2 + iPr (w + a Gr u_0)] q + Gr Pr (u + w Dt_0) \quad (18d)$$

$$Dw + iau + ibv = 0 \quad (18e)$$

with $L = k^2 + i(w + a Gr u_0)$ and $k^2 = a^2 + b^2$. We have also introduced the Grashof number: $Gr = Ra/Pr$. The boundary conditions are

$$\text{at } z = 0: u = v = w = q = 0, \quad (19a)$$

at $z = 1$: $w = 0$, $Dq + Bi\ q = 0$, (19b)

and $Du + iaWq = 0$, $Dv + ibWq = 0$ (19c)

The differential system (18) takes a simple form when attention is restricted to stationary modes ($w=0$) in the form of rolls with their axis along the streamwise direction ($a=0$). However, as in the general case the eight-order ordinary differential system is solved using a numerical procedure often used in the context of hydrodynamical stability [4]. Following this approach, the solution is sought as the superposition of four linearly independent solutions, each of them satisfying the boundary conditions at $z=0$. These solutions are constructed by a Runge-Kutta scheme of the fourth order with an appropriate step size. Satisfaction of the boundary conditions at $z=1$ leads to a homogeneous algebraic system which has non trivial solutions under the requirement of a vanishing determinant. For assigned values of the parameters Bi , W and Pr this leads to a characteristic equation of the type

$$F(k, w, Gr) = 0. (20)$$

The marginal stability curves are obtained by keeping the wave number k at a fixed value and determining the values of w and Gr for which the real part and the imaginary part of (20) vanish simultaneously.

For various values of the Biot number the neutral modes of instability have been identified when the parameter W is varied. The results are presented in the next section for the stationary and the oscillatory modes.

V. RESULTS AND DISCUSSION

A. Stationary modes

The existence of stationary modes as the preferred modes of instability for buoyant-thermocapillary flows has been previously reported in two circumstances. The first case was found by Smith and Davis [4] when analyzing the stability of the “linear flow“ driven by pure thermocapillary forces. The second case was found by Gershuni et al. [8] when analyzing the stability of buoyant-thermocapillary flow confined between perfectly conducting rigid-free horizontal surfaces. In both of these cases, the occurrence of stationary modes was associated with Prandtl number values larger than one, and also with a vertical temperature profile which is as a whole [4] or by part [8] destabilizing in the sense of Marangoni or Rayleigh-Bnard instability. Before presenting our results we shall mention that some peculiarities of these earlier models like consideration of a linear flow [4] as well as the choice of a conducting free surface [8] were irrelevant for an appropriate description of the experimental configuration presented in [15].

In this sub-section we consider the onset of stationary ($w=0$) longitudinal

rolls ($a=0$) for which the set of Eqs. (18) reduces to

$$(D^2 - b^2)u = GrwDu \quad (21a) \quad (D^2 - b^2)2w = b^2 q \quad (21b)$$

$$(D^2 - b^2)q = GrPr (w Dt_0 + u) \quad (21c)$$

with the boundary conditions

$$u = w = Dw = q = 0 \text{ at } z = 0 \quad (22a)$$

and at $z = 1$

$$w = Du = 0 \quad (22b)$$

$$Dv + ibWq = 0 \quad (22c)$$

$$Dq + Bi q = 0 \quad (22d)$$

Solutions were sought for values of the Biot number in the range $1 \leq Bi \leq 100$, and for values of W in the range $0 < W < 8$. We must notice that due to a different choice of length scale in Ref. 8 ($h/2$ instead of h) the parameter W used by these authors differs from ours by a factor 4. The higher value considered in [8] corresponds to $W=2.5$ in our notation.

We have found that for a given value of the Biot number the stationary longitudinal rolls are the preferred mode of instability provided that W does not exceed a certain value W_0 which is an increasing function of the Biot number. Representative neutral stability curves are drawn in Fig. 2 for two different values of W . The stability boundary $Gr(b)$ consists of two intersecting branches of a nearly parabolic shape, each of them having a local minimum at a given value of the wave number. For $Bi=10$ and $W=0.03$, the first minimum $Gr_1=870$, which is also the absolute minimum occurs on the left branch for the critical value of the wave number $bc=b_1=7.0$ while a secondary minimum $Gr_2=999$, occurs on the right branch for a higher wavenumber value $b_2=11.5$. For a higher value $W=0.1$, the situation is just the opposite: the absolute minimum $Gr_2=541$ occurs on the right branch for a critical value of the wave number $bc=b_2=13.1$ while a higher minimum $Gr_1=672$, occurs on the left branch at a lower value of the wavenumber $b_1=6.7$. Keeping the Biot number fixed, one can always find a value $W=W_{12}$ for which the two minima are equal $Gr_1=Gr_2$ and for $W < W_{12}$ instability occurs at $Gr_c=Gr_1$. For $Bi=10$, this value is $W_{12}=0.047$. However, anticipating on the results of the next section we shall mention that the value W_{12} is higher than the value W_0 at which unsteady modes become more critical. Therefore, the modes associated to the second minimum at b_2 are never critical and if we mention their existence this is because they have spatial properties very similar to that of the oscillatory modes we shall discuss in the next section.

The two stationary modes not only differ by the value of their respective

horizontal wavenumber but also by the vertical dependence of the eigenfunctions $w(z)$ which are drawn in Fig. 3 for two different values of W taken on both sides of W_{12} . It is shown that for $W < W_{12}$, $w(z)$ reaches its maximum amplitude in the lower half part of the layer while for $W > W_{12}$, $w(z)$ only takes substantial values in a narrow region confined near the free surface where it reaches its maximum value. Thus, two types of stationary modes with very different spatial properties have been identified. In fact these two types of modes are closely related to thermal modes of instability which have been already but separately identified in [4] and [8]. Those modes corresponding to the lower value of the wave number are connected to the ‘‘Rayleigh modes’’ which have been previously identified by Gershuni et al. [8] and involve classical Rayleigh-Bnard instability mechanism. Rayleigh modes can develop in regions where the basic temperature profile $t_0(z)$ is unstably stratified ($t_0' < 0$). As already pointed out in section III, this arises in two sub-layers adjacent to the lower and upper surfaces. The heights of these sublayers denoted hereafter h_B and h_T (for bottom and top) evolve with the parameters W and Bi . The most important evolution concerns the variation with the Biot number since h_B and h_T shrink to zero when $Bi=0$ (see Fig. 4). Moreover, the height h_B of the lower sublayer remains always larger than the height h_T of the upper sublayer. If we consider local Rayleigh numbers related respectively to the lower and upper sublayers they can be expressed approximately by

$$Ra_B \gg Dt(0)h_B^4 \text{ and } Ra_T \gg Dt(1)h_T^4$$

where we have assumed that the destabilizing temperature gradient in each sublayer is constant and equal respectively to its value at $z=0$ and at $z=1$. Since $Dt(0)=Dt(1)$ and $h_B > h_T$ then $Ra_B > Ra_T$. Though it is known that the critical value Ra_c of the Rayleigh number is sensitive to the choice of thermal and mechanical boundary conditions [15] we shall still assume that the value of Ra_c is the same in the two sublayers. Thus, the critical conditions will be reached at first in the lower sublayer and the instability will develop on a length scale of order h_B . However, owing to penetrative convection one can expect that the vertical extent of instability will be slightly greater than h_B but will not exceed $h/2$. This explains why the critical wavenumber associated with this type of instability has a characteristic value around $bc=6$, instead of $bc=3$ when Rayleigh instability spreads over the whole height of a layer bounded by conducting surfaces.

The second type of stationary modes is characterized by a larger value of the critical wave number indicating that the length scale of the instability is

related to the height hT of the upper sublayer. This region bounded above by a free-surface is susceptible to become unstable towards either Marangoni instability or Rayleigh-Bnard instability. The relative importance of the two mechanisms which are in competition in the upper part of the fluid layer depends on the boundary conditions (22c) and (22d). Inspection of Eq. (22c) shows that Marangoni instability will manifest itself through the coupling between the tangential stress and the surface temperature disturbance. Therefore, if either $W=0$ (no thermocapillary effects) or $q=0$ (conducting free-surface) Eq. (22c) reduces to a stress-free condition and the only destabilizing mechanism is Rayleigh-Bnard instability. This is the reason why Gershuni et al. [8] in their stability analysis of buoyant-thermocapillary flows ($W \neq 0$) with a conducting free-surface ($q=0$) never found Marangoni modes of instability. On the contrary, Smith and Davis [4] analysed the stability of pure thermocapillary flows (W) with a thermal boundary condition on the top free-surface alike Eq. (22d). These authors, who considered two different basic states, have identified a thermal mode of instability of the Marangoni type when dealing with a linear basic flow associated with a vertical temperature profile describing heating from below. It is likely that the same kind of instability is acting in the upper part of the liquid layer when there is an inverse-S-shaped temperature profile as in the present case. Moreover, in this upper region whose height is hT the velocity profile can be locally approximated by a linear flow (see Fig. 1) which enhances the similarity with the configuration of Smith and Davis [4]. However, it remains differences between their configuration and the present one which prevent from quantitative comparison between the corresponding results. The main difference concerns the mechanical boundary conditions on the bottom surface which is rigid in [4] while in the present case the upper sublayer is not bounded below and it is allowed for convective motion to penetrate deeper in the fluid. The large values of the critical wave number associated with Marangoni modes can be understood once the wavelength is rescaled with hT instead of h .

B. Oscillatory modes

For a fixed value of the Biot number while varying the parameter W , we have found a value W_0 above which the preferred modes of instability are oscillating at a frequency $\omega \neq 0$. These unsteady modes exist in the form of either longitudinal waves ($a=0, b \neq 0$), oblique waves ($a \neq 0, b \neq 0$) or transversal waves ($a \neq 0, b=0$). These oscillatory modes are reminiscent of the hydrothermal waves identified by Smith and Davis [4] in the case of a pure thermocapillary basic flow and of the traveling rolls found by Parmentier et

al. [9] in the case of a buoyant-thermocapillary basic flow and a zero value of the Biot number. The oscillatory modes are characterized by an overall wavenumber $k=(a^2+b^2)^{1/2}$ and by an angle of propagation $\pm F$ with respect to the negative x-axis defined as

$$F = \tan^{-1}(b/a)$$

Before going further we shall emphasize on the differences between our notations and those used in Refs. 4 and 9. In our case, the base flow velocity at the interface is directed towards the negative x-axis, contrary to what happens in [4, 9]. Moreover, the frequency w introduced in Eq. (17) is the opposite of the quantity called s in [4] and s in [9]. Thus, F has the same meaning here than in [4] and is related to the complementary angle $Y=1/2\pi - F$ also introduced in [4].

A typical stability diagram drawn in Fig. 5(a) for $Bi=10$ and $W=0.2$, shows the relative position of the neutral curves $Gr(b)$ for both stationary and oscillatory modes. The neutral curve for longitudinal waves ($a=0$, $w^1=0$) branches off from the neutral curves for stationary modes at a point corresponding to a value b_0 of the wavenumber. For $b < b_0$ longitudinal waves are more critical than stationary modes. For the whole range of wavenumbers b considered in Fig. 5(a) there are also oblique waves ($a^1 \neq 0$) which are the most unstable modes for the choice of parameters corresponding to Fig. 5(a). Moreover, it is shown on Fig. 5(a) that for large values of b the neutral curves for oblique waves are found in close vicinity of the stationary neutral curve for the Marangoni modes. The neutral curves for oblique waves exhibit strong deformations when increasing the value of a . For small and moderate values of a these neutral curves exhibit two local minima [Fig. 5(b)] at finite values of b , while for large values of a there is only a minimum which moves towards b_0 , as a increases [Fig. 5(c)].

Global minima of the Grashof number $Gr(a, b)$ were found by minimization with respect to both a and b . Under the conditions of Fig. 5(a), two global minima have been found. In this case, the first minimum which is also the absolute minimum occurs for $a=1.7$ and $b=3.1$, leading to a critical value of the wave number $kc=3.5$ and an angle of propagation $Fc=61^\circ$. The corresponding complex eigenfunction $w(z)$ was drawn on Fig. 6(a) showing that its amplitude which is maximum in the upper part of the fluid layer then decreases smoothly towards the bottom. A secondary global minimum was found for $a=7.5$ and $b=2.6$ leading to $k=7.9$ and an angle of propagation $F=81^\circ$. The corresponding complex eigenfunction $w(z)$ drawn on Fig. 6(b) only takes appreciable values in a narrow region confined near the free

surface as it was also the case for the stationary modes of the Marangoni type found in the previous subsection (see also Fig. 3).

When W is varied the existence of two distinct global minima is assessed providing that $W > W_B$, where W_B is the value for which the two minima coalesce. Another value of W of great importance is the value W_E for which the Grashof numbers corresponding to the two global minima are equal. For $Bi=10$, the value $W_E=0.17$ corresponds to an exchange of stability between two types of oscillatory modes. This exchange is accompanied by an abrupt change in the critical value of the characteristic parameters as shown in Fig. 7 for the wavenumber k_c , the angle of propagation F_c and the frequency w_c .

The situation depicted above for $Bi=10$, is that which prevails when the value W_E is larger than the value W_B . When decreasing the value of the Biot number, W_E becomes closer to W_B and for $Bi=7$ the two values coincide: $W_E=W_B$. This results in a continuous change of the critical parameters as shown on Fig. 8 for k_c , F_c and w_c . Though the curves plotted in Fig. 8 are continuous there is a break in their slope at the branching point $W=W_B$ where the second global minimum appears and becomes at once the most critical ($W_E=W_B$). For $Bi < 7$, the neutral curves exhibit only one minimum and the curves k_c, F_c and w_c versus W are smooth and continuous as shown in Fig. 9 for $Bi=1$.

From the above discussion, it results that for $Bi > 7$, a distinction has to be made between two types of oscillatory modes which differ by the order of magnitude of their critical parameters k_c , F_c and w_c and by the vertical extent on which the perturbed quantities take appreciable values. We shall label modes of type I those modes characterized by small values of the wavenumber ($3 < k < 5.5$), low values of the frequency ($w < 100$) and a large range of variation of the angle of propagation ($50^\circ \leq F \leq 120^\circ$, for $Bi=10$) when W is varied. We shall label modes of type II those modes characterized by higher values of the wavenumber ($5.5 \leq k \leq 8$), higher values of the frequency ($100 \leq w \leq 200$) and a smaller range of variation of the angle of propagation ($0^\circ \leq F \leq 20^\circ$, for $Bi=10$) when W is varied. The pure transversal waves ($F=0^\circ$) have been included in modes of type II.

The behavior found for $Bi=10$, remains qualitatively true for larger values of the Biot number, the main feature to be reported is that the domain of existence of oscillatory modes of type II increases with the value of Bi . In the $\{W, Bi\}$ plane the states diagram drawn in Fig. 10, shows the domain of existence of the different modes.

The Biot number being fixed, we shall summarize the sequence of tran-

sitions when W is increased above the threshold value W_0 for the onset of oscillatory instability. Just above W_0 pure transverse rolls are predicted to occur which evolve towards oblique rolls having a small angle of propagation. Further increasing W , the angle of propagation increases either continuously ($W_E=W_B$) or by an abrupt transition ($W_E>W_B$).

At this stage of the discussion it is worthwhile to recall the results of Parmentier et al. [9] who analyzed the stability of buoyant-thermocapillary flow in an infinite fluid layer bounded by two adiabatic surfaces. For the particular case $Pr=7$, they reported on two types of oscillatory modes they labelled (a) and (b) and which are characterized by different angles of propagation. Modes of type (a) occur in microgravity conditions ($0<Ra<335$) and are associated with small values of the critical angle of propagation $0\leq Y_c\leq 30^\circ$, and a negative phase speed $c=w/k$ in the range $-20<c<0$, (the time scale being h^2/k). In the case of a pure thermocapillary return basic flow ($Ra=0$ or W), Smith and Davis [4] have found that oblique hydrothermal waves with $Y_c=27^\circ$ for $Pr=6.8$ are the preferred mode of instability with $kc=2.6$ and $c=0.059$ (the time being scaled with h/gbT). Due to a different choice of notations this is the complementary angle $1^\circ - Y_c$ which must be compared with our results shown in Figs. 7(b)-8(b) and in Table 1. In our analysis the situation which better approaches the limit of a pure thermocapillary flow (W) between adiabatic horizontal surfaces ($Bi=0$) corresponds to $Bi=1$ and $W=8$. For this set of parameters the values of the critical parameters $kc=2.6$ and $wc=6$, presented in Table 1, are in agreement with the corresponding values found in [4] and in [9] for modes (a), while $Fc=133^\circ$, or $Y_c=1^\circ - Fc=47^\circ$, is larger than the value found by these authors. The modes of type (b) are the preferred modes of instability when buoyancy effects are in competition with thermocapillary effects and the angle of propagation has a larger range of variation: $0^\circ\leq Fc\leq 110^\circ$, while the positive phase speed takes large absolute values ($102<c<104$). For modes of type (b) the definition of the angle of propagation in [9] coincides with Fc , being the angle between the direction of propagation and the surface velocity $U_0(z=1)$. When buoyancy is dominant ($Ra>4\times 10^3$) the preferred mode of oscillatory instability found by Parmentier et al. [9] are transverse rolls with $Fc=0$. Just above the threshold for oscillatory instability $W>W_0$ we also found that transversal waves are the most critical, becoming later oblique waves. In the range W_0 , a comparison has been made between our results obtained for $Bi=1$, and the results of Parmentier et al. [9] corresponding to $Bi=0$ showing a qualitative agreement with modes of type (b). However it must be kept in mind that

the model of Parmentier et al. [9] is not an exact limiting case of our model. Even in the limit $Bi \rightarrow 0$, a different choice of boundary condition for the temperature disturbance on the lower rigid surface, which is conducting in our case while it is adiabatic in their case, prevents from a full identification of the two models.

VI. COMPARISON WITH EXPERIMENTS

The experimental results by Daviaud and Vince [15] have shown that the transition between stationary and traveling waves occurs for a value $W_0=1$ when the width of the container is $L_x=1\text{cm}$ and for $W_0=0.36$ when $L_x=2\text{cm}$. Our theoretical results yield the same order of magnitude for W_0 only if large values of the Biot number are considered since it was found that $W_0>0.36$ when $Bi>70$ (Fig. 10). It is unlikely that such large values for Bi fit in with the experimental thermal boundary conditions. It must be pointed out that the experimental value of W_0 is lowered when the width of the container is increased. Thus, one can argue that the lateral confinement in experiments stabilizes the stationary modes and delays the onset of oscillatory instability.

A. Stationary modes

The stationary longitudinal rolls which appear in experiments for large values of the height of fluid are characterized by a wavelength which is proportional to h , leading to a non dimensional wave number $b \approx 6$ in fair agreement with our theoretical findings. The stationary modes appear above a threshold value of the temperature difference which is plotted in [15] for different values of h . An estimate of the critical Grashof number can be made provided the horizontal gradient is assumed to be constant over the whole width of the container and equal to $bT=(T_1-T_0)/L_x$. This is far from to be true since the respectively ascending and descending flows near the hot and cold lateral walls modify the horizontal thermal gradient which is larger near the vertical walls than at mid-width of the container where our model is valid. The results obtained by Daviaud and Vince [15] for $0.9>W>0.3$, yield critical Grashof numbers in the range $2000<G_{rc}<7800$. Knowing that the critical value of bT is overestimated in experiments, this values must be divided by a factor 3 to 5 before comparison. Theoretical values of G_{rc} having the same order of magnitude are found if small values of the Biot number are considered but in that case the transition between stationary and oscillatory modes occurs at a value W_0 much lower than the corresponding experimental value. For instance, when $Bi=6$, stationary modes are critical provided that $W_0<0.03$ with a critical Grashof number $G_{rc}=1016$, when $W_0=0.03$.

B. Oscillatory modes

The traveling waves which appear in experiments [15] for small values of the height of fluid are characterized by an angle of propagation $Y \approx 80^\circ$ (or equivalently: $F \approx 100$), which strongly differs from the value $Y \approx 27^\circ$ found by Smith and Davis [4] when buoyancy was neglected and for $Pr=6.8$. It is worthwhile to notice that the experimental value of Y does not seem to depend on the value of W . Since the analysis of Parmentier et al. [9] performed in the case of adiabatic horizontal boundaries and for $Pr=7$, it is known that the addition of gravity allows for a larger range of variation of the angle of propagation which increases from $F=0$ to $F \approx 100$ when the Rayleigh number decreases from $Ra=4 \times 10^3$ to $Ra \approx 400$. It was shown in [9] that at $Ra=365$, the system undergoes a transition with an abrupt change of the angle of propagation which strongly decreases. In the present study where the thermal boundary conditions are different we have found two types of oscillatory modes, named I and II, which share some common features with the modes (a) and (b) found by Parmentier et al. [9] but cannot be totally identified with them. The main difference concerns the asymptotic value of F when W is large. For realistic values of the Biot number ($1 < Bi < 10$), we always found $F > 100$ when $W > 1$, a result which better agrees with the experimental value of Daviaud and Vince [15] than did previous theoretical results [4,9]. The experimental wavelengths measured in [15] vary in the range $4h < l < 5.5h$, thus corresponding to values of the wavenumber $1.14 < kc < 1.57$, smaller than the theoretical values: $2.5 < kc < 3$, found for $W > 1$ and $1 < Bi < 10$. The non dimensional frequency $w = (2\pi/t)(h^2/\nu)$, can be deduced from the plot of the period t drawn in [15]. For values of the height: $1\text{mm} < h < 2\text{mm}$, or correspondingly $2 < W < 8$, this leads to a nearly constant value $w \approx 10$, which can easily be recovered as a result of our calculations. For instance, when $Bi=1$ and $W=2$ we have found $w=10.9$ and $Grc=26$. The experimental values of the critical Grashof number range from $Grc=15$ when $W=8$ ($h=1\text{mm}$) to $Grc=200$ when $W=2$ ($h=2\text{mm}$). Remembering that the experimental values of the thermal gradient are overestimated by at least a factor 3, these values of Grc are compatible with our theoretical findings listed in Table 1. The main difference between our results and the experimental observations by Daviaud and Vince [15] concerns the existence of oscillatory modes having a small angle of propagation (nearly transversal waves). These modes which are predicted to exist just above the onset of oscillatory modes (Fig. 10) have never been observed in [15]. The reason for such a discrepancy may be due to the finite extent of the container in the direction of the thermal gradient. In the opposite geometry, when Lx is large compared to the extent in

the y-direction, Villers and Platten [12] reported on the existence of spatio-temporal states which are multi-cellular in the x-direction but which cannot be simply identified to traveling waves. In a configuration where $L_x \ll L_y$, evidence of transversal structures have been reported in [13].

VII. CONCLUSIONS

The aim of this study was to analyse the linear stability of coupled buoyant-thermocapillary-driven flow induced by a horizontal temperature gradient in fluid layers of infinite horizontal extent. The thermal boundary conditions on the top and bottom horizontal surfaces were taken non symmetrical; the rigid bottom wall was maintained at a temperature varying linearly along the direction of the thermal gradient while a mixed thermal boundary condition was chosen to describe heat transfer at the upper free surface. The critical modes have been determined when the Biot number and the ratio of Marangoni number to Rayleigh number ($W = Ma/Ra$) were varied. When the Biot number is large and buoyancy effects are dominant the system is prone to destabilize towards stationary modes in the form of pure longitudinal rolls. When the Biot number is small and thermocapillary effects are dominant the system is prone to destabilize towards oscillatory modes in the form of oblique waves. For intermediate values of the Biot number the system undergoes transitions between different states as W varies. When buoyancy effects are dominant ($W < W_0$) the preferred modes of instability are stationary longitudinal rolls. Just above the value W_0 , the most critical modes are oscillatory modes in the form of nearly transversal waves with a small angle of propagation ($0^\circ < \theta_c < 20^\circ$). When W increases and reaches a value W_E , a second transition occurs which is characterized by a jump in the behavior of the critical parameters. For $W > W_E$, the angle of propagation increases and even reaches the value $\theta_c \approx 90^\circ$, whereas the critical wavenumber and frequency are lowered.

Our theoretical results have been compared with the experimental observations reported by Daviaud and Vince [15]. We have been able to reproduce the transition they observed between stationary and oscillatory modes when the height of fluid is increased. We believe that our choice of thermal boundary conditions was decisive in order to exhibit this transition. However, it was not possible to fit simultaneously the value of the Biot number and the value of W_0 at which the transition occurs. This may be due to the finite extent of the container used in [15] which stabilizes longitudinal stationary rolls or to a wrong estimate of the Biot number. When the stationary modes are critical, we have shown that our model yields a critical value of the wave

number which is in good agreement with the experimental measurements. The predicted value of the critical Grashof number is lower than in experiments; the discrepancy is now well understood being due to an overestimate of the horizontal thermal gradient in experiments. As a result of our calculations we also found two types of oscillatory modes which differs by the value of their critical parameters. Modes of type II which are predicted to occur just above the transition between stationary and oscillatory modes have never been detected in experiments; they are characterized by a small value of the angle of propagation and large values of both wavenumber and frequency. Modes of type I become critical after a second transition has occurred at $W=WE$ which is accompanied by an abrupt change in the critical parameters; they are characterized by larger values of the angle of propagation and smaller values of the wavenumber and frequency. The properties of modes I better suit with the experimental findings even though it remains a discrepancy about the wavelength which is twice larger in experiments.

In earlier studies, when a transition between stationary and oscillatory instability modes was reported it resulted from changing the Prandtl number. For instance, Smith and Davis [4] have found a transition between hydrothermal waves and longitudinal rolls for $Pr=0.82$ when analysing the stability of a linear flow driven by thermocapillarity and confined between adiabatic horizontal surfaces of infinite extent. With the addition of buoyancy effects and different thermal boundary conditions (conducting surfaces) Gershuni et al. [8] have shown that the most dangerous mode of instability is either oscillating ($Pr<1$) or stationary ($Pr>1$). When considering the instability of axisymmetric thermocapillary flow in a cylindrical bridge, Wanschura et al. [20] reported that the primary bifurcation is stationary if $Pr\ll 1$, while the basic state becomes unstable to a pair of hydrothermal waves when $Pr=4$. These authors anticipated that the stationary instabilities observed in straight cavities [11, 15] are due to the small Prandtl number mechanism of instability. This assertion is in contradiction with the results found in [4, 8] and with our own calculations performed with $Pr=7$.

Smith [19] has shown that instability mechanisms in dynamic thermocapillary liquid layers are different according to the value of the Prandtl number. In the limit of small Prandtl number as well as in the opposite limit of large Prandtl number, Smith [19] clearly showed how the interaction between the disturbance convection field and the basic-state temperature and velocity field determines the instability mode. The distinction he made between the return flow and the linear flow basic-states often hides the difference between

the associated temperature profiles which are respectively stabilizing in the first case and destabilizing in the latter case. We believe that when thermal instability mechanisms are dominant (for moderate and large Prandtl numbers) the role of thermal boundary conditions should deserve a better attention in future work. The need for more experimental data is welcomed, especially to elucidate the role of the geometrical configuration in the selection of a particular mode of instability.

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FIGURE CAPTION

Figure 1: The basic states for $W=1$: (a) velocity profile; (b) temperature profiles: (—) $m=0$, (...) $m=0.5$ and () $m=1$.

Figure 2: Neutral stability curves for stationary modes and $Bi=10$: () $W=0.03$, (...) $W=0.1$.

Figure 3: Vertical dependance of the eigenfunctions velocity component $w(z)$ for $Bi=10$: () $W=0.03$, (...) $W=0.1$.

Figure 4: Evolution versus Bi of the heights hB and hT corresponding to the lower and upper sublayers unstably stratified in temperature, for $W=1$: () hB , (...) hT

Figure 5: Neutral stability curves for oscillatory modes, for $Bi=10$ and $W=0.2$; (a): (- - -) $a=0$, (—) $a=1$, (...) $a=5.5$, the full lines are neutral curves for the stationary modes; (b): (- - -) $a=1$, (—) $a=1.7$, (...) $a=2.5$; (c): (- - -) $a=6.5$, (—) $a=7.5$, (...) $a=8.5$.

Figure 6: Vertical dependance of the complex eigenfunctions velocity component $w(z)$ for $Bi=10$ and $W=0.2$: (a) $kc=3.5$, () real part, (...) imaginary part; (b) $kc=7.9$.

Figure 7: Critical values of the characteristic parameters versus W for $Bi=10$: (a) wavenumber kc ; (b) angle of propagation Yc ; (c) frequency wc . The two vertical dashed lines indicate respectively the branching point at $WB=0.14$, and the exchange of stability at $WE=0.17$. The critical behavior is represented by full lines.

Figure 8: Critical values of the characteristic parameters versus W for $Bi=7$: (a) wavenumber kc ; (b) angle of propagation Yc ; (c) frequency wc . Here $WB=WE=0.105$. The critical behavior is represented by full lines.

Figure 9: Critical values of the characteristic parameters versus W for $Bi=1$: (a) wavenumber kc ; (b) angle of propagation Yc ; (c) frequency wc .

Figure 10: States diagram showing the domain of existence of the different critical modes in the (Bi, W) plane. In the region $Bi < 7$, the transition between oscillatory modes of type I and II can no longer be made on the basis of an abrupt change of the critical parameters kc , Yc , wc .

TABLE CAPTION:

Table1: Values of carateristic parameters for two values of Biot number and large W values.

Bi110

W125812510

k2,552,592,622,622,942,852,782,69

F101,31117,55133,45133,4599,80108,4120,30120,30

w18,5510,906,156,0017,2211,716,516,10

Gr55,1025,998,955,4172,4635,1212,866,16